

# Separating the natural and anthropogenic air-sea flux of CO<sub>2</sub>: The Indian Ocean

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[1] We estimate the natural and anthropogenic components of the air-sea flux of CO<sub>2</sub> in the Indian Ocean. The increase in atmospheric CO<sub>2</sub> driven by human activity has caused the air-sea CO<sub>2</sub> disequilibrium, and consequently the flux, to increase significantly over the industrial era. We estimate the flux in the year 1780 to be approximately 0.2 Gt/yr, increasing by 0.26 Gt/yr to 0.5 Gt/yr in 2000. The estimate of the natural (preindustrial) flux is highly sensitive to uncertainties in modern-day CO<sub>2</sub> disequilibrium measurements. By contrast, the estimate of the anthropogenic flux is only weakly sensitive to these measurements. **INDEX TERMS:** 4806 Oceanography: Biological and Chemical: Carbon cycling; 4572 Oceanography: Physical: Upper ocean processes; 4568 Oceanography: Physical: Turbulence, diffusion, and mixing processes; 4808 Oceanography: Biological and Chemical: Chemical tracers. **Citation:** Hall, T. M., and F. W. Primeau (2004), Separating the natural and anthropogenic air-sea flux of CO<sub>2</sub>: The Indian Ocean, *Geophys. Res. Lett.*, 31, L23302, doi:10.1029/2004GL020589.

## 1. Introduction

[2] The ocean plays a major role in the perturbed carbon cycle, but quantifying the uptake and ocean inventory of anthropogenic carbon is difficult, and considerable uncertainties remain. The fractional magnitude of the perturbation is also uncertain, because the “natural” state of air-sea CO<sub>2</sub> flux prior to the industrial era is poorly known. The flux of carbon into the ocean is driven by the difference (“disequilibrium”) between the atmospheric CO<sub>2</sub> partial pressure ( $p\text{CO}_{2,\text{atm}}$ ) and the oceanic partial pressure ( $p\text{CO}_{2,\text{ocn}}$ ) that would be in equilibrium with the dissolved inorganic carbon (DIC) concentration of surface waters. Measurements of surface-water DIC have allowed estimates of the modern-day air-sea disequilibrium,  $\Delta p\text{CO}_2 \equiv p\text{CO}_{2,\text{atm}} - p\text{CO}_{2,\text{ocn}}$ , and therefore the total flux [Takahashi *et al.*, 2002], which is comprised of anthropogenic and natural (preindustrial) components. Prior to the industrial era the globally-averaged ocean was in approximate equilibrium with the atmosphere, but locally, the air-sea disequilibrium, and consequently the flux, was likely significant in many places. Oceanic entry and exit regions of carbon do not coincide, partly due to regionally varying transport to and from the surface and partly due to regionally varying biota, which consume DIC. Superposed on this background natural disequilibrium is the oceanic signal of increasing atmo-

spheric  $p\text{CO}_{2,\text{atm}}$  due to human activity. Rising  $p\text{CO}_{2,\text{atm}}$  in the presence of a finite air-sea equilibration time causes  $\Delta p\text{CO}_2$  to become more positive, driving an increased carbon flux into the ocean.

[3] Here we separate the natural and anthropogenic components of the CO<sub>2</sub> flux into the Indian Ocean. We employ the method of Hall *et al.* [2004] to compute the anthropogenic component, but generalized to include the dependence of the DIC perturbation on the preindustrial state. This generalization results in only a small change ( $\sim 2\%$ ) to the anthropogenic Indian Ocean carbon mass and uptake estimated by Hall *et al.* [2004]. A new feature of the analysis, however, is an estimate of the natural background flux.

## 2. Methodology

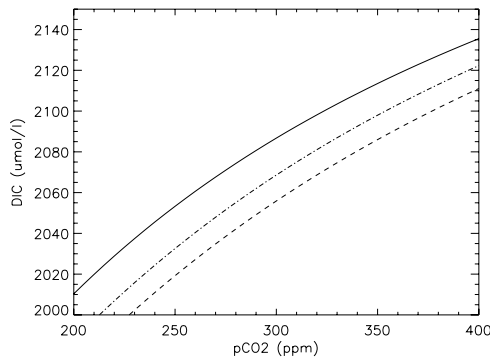
[4] The flux,  $F$ , of carbon across the air-sea interface is driven by the air-sea disequilibrium in  $p\text{CO}_2$ , and is comprised of natural and anthropogenic components. The anthropogenic component is

$$F'(t) = k\Delta p\text{CO}_2'(t) = k(p\text{CO}_{2,\text{atm}}'(t) - p\text{CO}_{2,\text{ocn}}'(t)) \quad (1)$$

where primes indicate anthropogenic quantities and  $k$  is the exchange coefficient, dependent on wind-speed and solubility. The industrial era is assumed to begin in 1780 ( $p\text{CO}_{2,\text{atm}}(1780) \approx 280 \mu\text{atm}$ ), so that  $F'(1780) = 0$ .

[5] Equation (1) has two unknowns,  $F'$  and  $p\text{CO}_{2,\text{ocn}}'$ . (The atmospheric perturbation,  $p\text{CO}_{2,\text{atm}}'$ , is determined from historical records and ice-core samples [Etheridge *et al.*, 1998]. Present-day values of the time-series we use closely match observations from Indian Ocean sites.) To close the system Hall *et al.* [2004] employ a second, independent relationship:  $F'(t) = dM'/dt$ , where  $M'$  is the mass of anthropogenic carbon in a constant-density (isopycnal) water volume receiving the flux. (The analysis is performed separately on each isopycnal volume in a range of densities spanning the Indian Ocean thermocline, the dominant receptacle of anthropogenic carbon, and the results summed to obtain a total flux.)  $M'$ , in turn, is expressed in terms of the history of DIC' in surface waters (outcrops) of the isopycnal volumes and the transit-time distribution,  $G_V(t)$ , which relates the mean concentration over volume  $V$  of any tracer to the tracer's concentration history on the outcrop. Combining, one has

$$F'(t) = V \frac{d}{dt} \int_{1780}^t dt' \text{DIC}'(t') G_V(t - t'). \quad (2)$$



**Figure 1.** Equilibrium DIC- $p\text{CO}_{2,ocn}$  relationship for three sample densities:  $\sigma_0 = 26.7$  (solid),  $\sigma_0 = 26.1$  (dot-dash), and  $\sigma_0 = 25.5$  (dash).

Equating (1) and (2) provides an integral equation for  $\text{DIC}'(t)$ , if a relationship between  $p\text{CO}_{2,ocn}'$  and  $\text{DIC}'$  can be obtained and an estimate of  $\mathcal{G}_V(t)$  is available.

[6] By definition, DIC and  $p\text{CO}_{2,ocn}$  are related by the equilibrium inorganic carbon system, and we write  $\text{DIC} = f(p\text{CO}_{2,ocn})$ . Given one (and knowledge of  $T$ ,  $S$ , and alkalinity) the other can be computed. This equilibrium relationship is nonlinear. As  $\text{CO}_2$  dissolves in sea-water the pH decreases, so that the change in DIC is not proportional to the change in  $\text{CO}_2$ . Consequently, the rate of increase of DIC with  $p\text{CO}_{2,ocn}$  is progressively slower as  $p\text{CO}_{2,ocn}$  increases. This is the well-known buffering effect and is summarized by the Revelle factor,  $\text{Re} = (d\text{CO}_2/\text{CO}_2)/(d\text{DIC}/\text{DIC})$ , which is typically in the range 8–15.

[7] Buffering has consequences for the relationship between the perturbation quantities  $p\text{CO}_{2,ocn}'$  and  $\text{DIC}'$  necessary to solve (1) and (2): the value of the natural background state affects the magnitude of  $\text{DIC}'$  in response to a perturbation  $p\text{CO}_{2,ocn}'$ . Figure 1 shows examples computed from the equilibrium inorganic chemistry model “CO2sys” [Lewis and Wallace, 1998] with alkalinity expressed as a function of salinity, as by Millero *et al.* [1998]. On the  $\sigma_0 = 26.7$  isopycnal an increase in  $p\text{CO}_{2,ocn}$  of 90  $\mu\text{atm}$  (i.e.,  $p\text{CO}_{2,ocn}' = 90 \mu\text{atm}$ ) starting at 200  $\mu\text{atm}$  results in  $\text{DIC}' = 70 \mu\text{mol/kg}$ , while the same 90  $\mu\text{atm}$  increase starting from 280  $\mu\text{atm}$  results in only  $\text{DIC}' = 46 \mu\text{mol/kg}$ . Upon equating (1) and (2) we write

$$\begin{aligned} & k(p\text{CO}_{2,atm}'(t) - (p\text{CO}_{2,ocn}'(t) - p\text{CO}_{2,ocn}(1780))) \\ &= V \frac{d}{dt} \int_{1780}^t dt' (f(p\text{CO}_{2,ocn}'(t')) - f(p\text{CO}_{2,ocn}(1780))) \\ & \cdot \mathcal{G}_V(t - t'), \end{aligned} \quad (3)$$

which makes explicit the fact that the preindustrial state must be specified.

[8] Unfortunately, the disequilibrium in the preindustrial era is not known. Takahashi *et al.* [2002], however, have made observationally based estimates of the total disequilibrium (natural plus anthropogenic) for the year 1995. We have averaged the annual-mean Takahashi values onto the outcrops of the Indian Ocean isopycnals (Figure 2). We separate the natural and anthropogenic disequilibrium components on each outcrop by iterating (3) for varying  $p\text{CO}_{2,ocn}(1780)$  until  $\Delta p\text{CO}_2(1995)$  matches the Takahashi

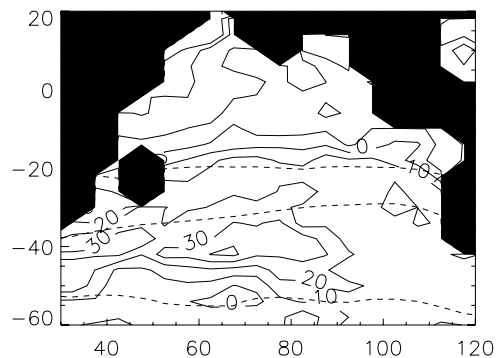
value within a small tolerance. At each step of the iteration (3) is solved numerically for  $p\text{CO}_{2,ocn}(t)$ , using  $\mathcal{G}_V(t)$  as described below. For  $k$  we use the estimates of Carr *et al.* [2002], based on QuickScat and SSM/I scatterometer data. Once  $p\text{CO}_{2,ocn}(t)$  is known  $\text{DIC}(t)$  (and thus  $\text{DIC}'$ ) can be computed from the equilibrium relationship, and, finally,  $F(t)$  can be computed from (2). In addition, we have the natural background flux  $F(1780) = k(p\text{CO}_{2,atm}(1780) - p\text{CO}_{2,ocn}(1780))$ .

[9] The  $\mathcal{G}_V(t)$  required for the calculation are derived from CFC data, as described in detail by Hall *et al.* [2004]. Briefly, CFC12 measurements in the Indian Ocean made during WOCE are used to constrain a two-parameter model of  $\mathcal{G}_V(t)$  for isopycnal volumes  $V$ .  $\mathcal{G}_V$  propagates tracer signals on the outcrop into the interior, as in (2), and has the interpretation as the distribution of transit times since water in  $V$  made last contact with the surface [e.g., Haine and Hall, 2002; Primeau, 2004]. The two parameters are the  $V$ -averaged Peclet number,  $Pe$ , and the mean transit time,  $\Gamma$ . A single datum (i.e., a  $V$ -averaged CFC12 concentration) cannot simultaneously constrain the two parameters. Instead, a family of  $\mathcal{G}_V$  is obtained, all of which are consistent with the CFC12 value. The high  $Pe$  limit, representing weak diffusive mixing, is equivalent to using a  $V$ -averaged CFC concentration age,  $\tau_{\text{CFC}}$ , as a lag time in the surface-water  $\text{DIC}'$  history, i.e.,  $\text{DIC}'_{\text{interior}} = \text{DIC}'_{\text{surface}}(t - \tau_{\text{CFC}})$ . Use of concentration ages in this way cause a positive bias in  $\text{DIC}'_{\text{interior}}$  estimates [e.g., Gruber *et al.*, 1996; Hall *et al.*, 2002]. In fact, Hall *et al.* [2004] for the Indian Ocean and Waugh *et al.* [2004] for the North Atlantic find that the low  $Pe$  limit (strong mixing) is most consistent with multiple tracers analyzed in combination, and we use this limit here. The low  $Pe$  limit results in about 30% less anthropogenic carbon mass in the Indian Ocean than the high  $Pe$  limit [Hall *et al.*, 2004].

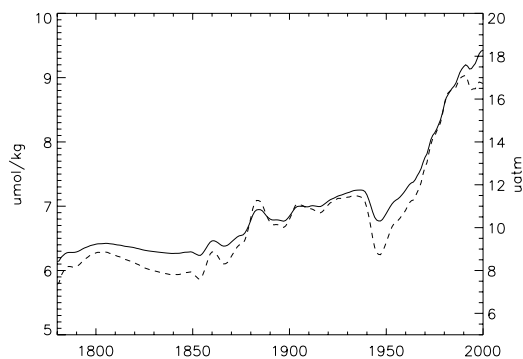
### 3. Results

#### 3.1. Preindustrial and Anthropogenic Fluxes

[10] Figure 3 shows the change in disequilibrium over the industrial era at the outcrop of the  $\sigma_0 = 26.5$  surface. We estimate  $\Delta p\text{CO}_2(1780) \approx 8 \mu\text{atm}$  for this water. The disequilibrium has increased rapidly since the 1950s, reaching about 18  $\mu\text{atm}$  in 2000. Other isopycnals have similar



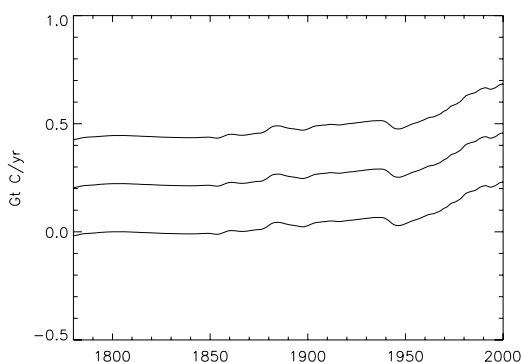
**Figure 2.** Distribution of total annual-mean Indian Ocean disequilibrium ( $\mu\text{atm}$ , solid) for, 1995 [Takahashi *et al.*, 2002]. Also shown are the three outcrops of  $\sigma_0$  isopycnals: 27, 25, and 23 from south to north (dash).



**Figure 3.** History of disequilibrium on  $\sigma_0 = 26.5$  expressed in terms of  $p\text{CO}_2$  (solid, right axis) and DIC (dash, left axis).

patterns, though most have smaller magnitude changes. Note that  $\Delta p\text{CO}_2(t)$  displays decadal variability, even though  $p\text{CO}_{2,\text{atm}}(t)$  increases monotonically. The evolution of disequilibrium is related to the time derivative of the carbon inventory (expression (3)), and is therefore more sensitive to the time derivative of  $p\text{CO}_{2,\text{atm}}(t)$ , which has decadal variability, than to  $p\text{CO}_{2,\text{atm}}(t)$  itself.

[11] We compute the flux into each isopycnal volume from its disequilibrium and sum over all the isopycnals to obtain the total CO<sub>2</sub> flux into the Indian Ocean. Figure 4 shows our best estimate of the total flux (solid curve), which uses the Takahashi disequilibrium data, as described above. The flux has increased from about 0.2 Gt/yr in 1780 to about 0.5 Gt/yr in 2000. The present-day total flux is in approximate agreement with the estimate of *Takahashi et al.* [2002] for the Indian Ocean north of 50°S. The anthropogenic flux is 0.26 Gt/yr in year 2000, and the anthropogenic inventory is 14.1Gt. These anthropogenic values are within a few percent of the estimates of *Hall et al.* [2004] (in their strong mixing limit), showing that the overall effect of including the dependency on the preindustrial state is small. (The effect is as large as 11% on certain isopycnals.) The small correction is well within the  $\pm 20\%$  uncertainty estimated by *Hall et al.* [2004] for their anthropogenic inventory and flux estimates. As in *Hall et*



**Figure 4.** Total carbon flux (Gt/yr) of into the Indian Ocean from 1780 to 2000. The center curve is derived using Takahashi disequilibrium measurements, while the upper (lower) curve is derived adding (subtracting) 5 ppm uniformly to the Takahashi values.

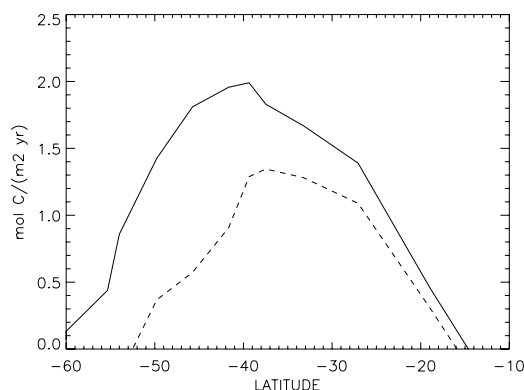
*al.* [2004], our anthropogenic flux and inventory estimates are about one third smaller than those of *Sabine et al.* [1999] and *McNeil et al.* [2003], due primarily to the allowance for mixing in the form of  $\mathcal{G}_V(t)$ .

[12] Also shown in Figure 4 are fluxes computed by adding  $\pm 5 \mu\text{atm}$  everywhere to the Takahashi disequilibrium, a conservative estimate of the uncertainty. The estimate of the natural background flux is highly sensitive to this uncertainty, and varies from  $-0.02$  Gt/yr to  $0.68$  Gt/yr with the  $\pm 5 \mu\text{atm}$  range. Importantly, however, the estimate of the anthropogenic flux component is only weakly sensitive to the disequilibrium uncertainty, ranging from  $0.25$  Gt/yr to  $0.26$  Gt/yr for  $\pm 5 \mu\text{atm}$ . Consequently the flux curves in Figure 4 are nearly parallel.

[13] Figure 5 shows the flux as a function of equivalent latitude for the years 1780 and 2000. (Equivalent latitude is the latitude that encompasses the same Indian Ocean area as the outcrop contours, and is close to true latitude because the outcrops are oriented approximately zonally.) The total flux peaks around 35°S to 40°S, but the anthropogenic component is largest further south. The 26.7 isopycnal, which has the most anthropogenic carbon per unit density, outcrops around 46°S.

### 3.2. Robustness of Anthropogenic Estimate

[14] The anthropogenic flux estimate is only weakly sensitive to the disequilibrium measurements because the equilibrium carbon chemistry is only weakly nonlinear. In the linear limit the perturbation quantities  $p\text{CO}'_{2,\text{ocn}}$  and  $\text{DIC}'$  would be related without any reference to the preindustrial state. In this limit (1) and (2) would provide two equations in two unknowns,  $F'$  and  $p\text{CO}'_{2,\text{ocn}}$ , which could be solved independent of any disequilibrium measurements. In fact, past studies have used a linearized equilibrium relationship to make CO<sub>2</sub> flux and inventory estimates [e.g., *Thomas et al.*, 2001]. (Even in the linearized case, an estimate of the natural flux requires disequilibrium measurements. The separation of anthropogenic and natural fluxes, however, is simpler: given  $p\text{CO}'_{2,\text{ocn}}$  from (1) and (2), one computes the change in disequilibrium over the industrial era,  $p\text{CO}'_{2,\text{atm}} - p\text{CO}'_{2,\text{ocn}}$ , which is subtracted from the present-day total disequilibrium measurement, leaving the natural component. This is converted to flux by multiplication with the exchange coefficient.)



**Figure 5.** Carbon flux ( $\text{mol C m}^{-2} \text{s}^{-1}$ ) versus equivalent latitude in the Indian Ocean estimated for year 2000 (solid) and year 1780 (dash).



[15] An additional feature contributing to the robustness of the anthropogenic estimate arises, in fact, from the nonlinearity (buffering effect) of the equilibrium relationship. Any method, including ours, that computes anthropogenic DIC by relating interior values to the surface-water history is only weakly sensitive to the anthropogenic changes in the  $p\text{CO}_2$  disequilibrium. The impact of any change in disequilibrium due to rising  $p\text{CO}_{2,\text{atm}}$  occurs through DIC (see expression (2)), and the change in DIC disequilibrium over the industrial era is smaller than the change in  $p\text{CO}_2$  disequilibrium. Define the DIC disequilibrium as  $\Delta\text{DIC} = \text{DIC}_{\text{eq}}(t) - \text{DIC}(t)$ , where  $\text{DIC}_{\text{eq}}(t)$  is the DIC that would be in equilibrium with  $p\text{CO}_{2,\text{atm}}(t)$ . The dashed line in Figure 3 shows  $\Delta\text{DIC}(1780) \approx 6 \mu\text{mol/kg}$  on the 26.5 isopycnal, reaching approximately  $9 \mu\text{mol/kg}$  in 2000, a much smaller change than the change in  $\Delta p\text{CO}_2$  (solid line). This is due to the buffering effect: While  $p\text{CO}_{2,\text{atm}}$  rises at an increasingly rapid rate, leading to an increasingly large lag of  $p\text{CO}_{2,\text{ocn}}$  (i.e., increasing disequilibrium), the rate of increase of  $\text{DIC}_{\text{eq}}$  with  $p\text{CO}_{2,\text{atm}}$  is progressively slower at higher  $p\text{CO}_{2,\text{atm}}$ , offsetting the tendency for increased disequilibrium in DIC. That is, DIC and  $\text{DIC}_{\text{eq}}$  cannot separate as easily as  $p\text{CO}_{2,\text{ocn}}$  and  $p\text{CO}_{2,\text{atm}}$ . In fact, a number of studies have neglected the change in disequilibrium in their estimates of anthropogenic flux and inventory [Gruber *et al.*, 1996; Sabine *et al.*, 1999; McNeil *et al.*, 2003; Thomas *et al.*, 2001], thereby allowing  $\text{DIC}'(t)$  to be computed entirely from equilibrium chemistry [Thomas *et al.*, 2001]. Due to the canceling effects of increased  $\Delta p\text{CO}_2$  and increased buffering this assumption appears to incur only modest error.

[16] Because the anthropogenic estimate is only weakly sensitive to changes in disequilibrium, it is also only weakly sensitive to uncertainty in the exchange coefficient,  $k$ . In the limit that equilibrium chemistry alone is sufficient to obtain  $\text{DIC}'(t)$  expression (2), which is independent of  $k$ , can be used alone to estimate  $F'$ . By contrast, the estimate of the natural flux component is directly dependent on  $k$  and, therefore, sensitive to its uncertainty.

#### 4. Summary and Conclusions

[17] We have estimated the natural and anthropogenic components of CO<sub>2</sub> flux into the Indian Ocean. In 1780, at the start of the industrial era, the flux was approximately 0.2 Gt/yr, and has increased by 0.26 Gt/yr to approximately 0.5 Gt/yr in the year 2000. The anthropogenic inventory in 2000 was 14.1 Gt. These anthropogenic flux and inventory estimates are marginally different ( $\sim 2\%$ ) from those of Hall *et al.* [2004], due to the inclusion here of the dependence on the preindustrial state. The estimate of the natural background flux is a new feature of this analysis. As a function of latitude the natural flux peaks around 35°S to 40°S, while the anthropogenic flux peaks around 46°S.

[18] The natural flux estimate is highly sensitive to the uncertainty in modern-day disequilibrium measurements

used in the analysis. We obtain a range of approximately 0.0 Gt/yr to 0.7 Gt/yr based on conservative estimates of the disequilibrium uncertainty ( $\pm 5 \mu\text{atm}$ ). By contrast, the anthropogenic component is weakly sensitive to this uncertainty, ranging only from 0.25 Gt/yr to 0.26 Gt/yr. The anthropogenic estimates are subject to the same  $\pm 20\%$  uncertainty estimated by Hall *et al.* [2004], and the analysis assumes that ocean transport is in steady-state, as do other analyses. However, the second-order sensitivity of our anthropogenic estimates to disequilibrium uncertainty is in stark contrast to the first-order sensitivity of anthropogenic flux estimates based directly on disequilibrium measurements.

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